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(71)Applicant : ASAHI GLASS CO LTD

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(72)Inventor : DOI TAKAO
WATABE TAKASHI
ONOGUCHI TATSUO
HAYASHI TOMOYOSHI

(54) IMPROVED ROOM-TEMPERATURE-CURABLE COMPOSITION

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a room-temp.-curable compsn. improved in depth curability and adhesiveness by incorporating a polymer having hydrolyzable silicon groups and at least one cure catalyst selected from among tin compds. into the same.

SOLUTION: This compsn. contains 100 pts.wt. polymer comprising a polymer (A) having hydrolyzable silicon groups represented by the formula: $-\text{SiXaR13-a}$ and/or a polymer (B) having a polyoxyalkylene main chain, 0.01-10 pts.wt. at least one cure catalyst selected from among tin compds., and a filler, a plasticizer, etc. The tin compds. include a compd. (K) represented by the formula: R22Sn(OZ)2 , a compd. (L) represented by the formula: $[\text{R22Sn(OZ)2}]_n\text{O}$, reaction product (M) of compd. K and a low-molecular compd. having a hydrolyzable silicon group, and a reaction product (N) of compd. L and a low-molecular compd. having a hydrolyzable silicon group. At least a part of polymer A is a polymer having a hydrolyzable silicon group of the formula wherein a is 3. In the formula, R1 is a monovalent org. group; R2 and Z are each a monovalent hydrocarbon group; X is a hydroxyl or hydrolyzable group; and a is 1-3.

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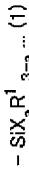
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CLAIMS

[Claim(s)]

[Claim 1] It is a polymer which has a hydrolytic silicon group expressed with a following formula (1), A room-temperature-curing nature constituent which uses as an essential ingredient at least one sort of tin compounds (K) in which some or all of this polymer is chosen from a polymer (A) which is a polymer which has a hydrolytic silicon group whose a of a formula (1) is 3, and a group which consists of following - (K-1) (K-4) as a curing catalyst.

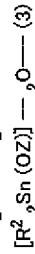
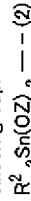


(R¹ shows a univalent organic group substitution of the carbon numbers 1-20, or unsubstituted among a formula (1), X shows a hydroxyl group or a hydrolytic basis, and a shows 1, 2, or 3,) however, when those R¹ may be the same, or may differ, when two or more R¹ exist, and two or more X exists, those X may be the same or may differ, tin compound (K)(K-1): — a tin compound expressed with a following formula (2).

(K-2): A tin compound expressed with a following formula (3).

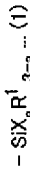
(K-3): (K-1) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.

(K-4): (K-2) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.



(R² is a univalent hydrocarbon group of the carbon numbers 1-20 among a formula (2) and (3), and Z is a univalent hydrocarbon group of the carbon numbers 1-20, or an organic group which has a portion which can form a coordinate bond to Sn by intramolecular.) two or more R² may be the same, or may differ, and two or more Z may be the same, or may differ.

[Claim 2] It is a polymer which has a hydrolytic silicon group expressed with a following formula (1), A room-temperature-curing nature constituent for which some or all of this polymer uses as an essential ingredient a tin compound (M) which consists of following (M-1) and/or (M-2) as a polymer (A) which is a polymer in which a of (1) has a hydrolytic silicon group which is 3, and a curing catalyst.



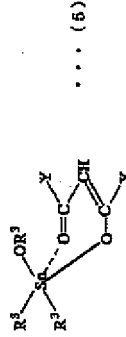
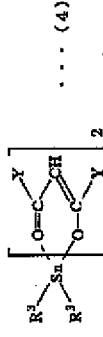
(R¹ is a univalent organic group substitution of the carbon numbers 1-20, or unsubstituted among a formula (1), X is a hydroxyl group or a hydrolytic basis, and a is 1, 2, or 3,) however, when * and those R¹ may be the same, or may differ from each other, when two or more R¹ exist, and two or more X exists, those X may be the same or may differ.

A tin compound (M): (M-1): An oxygenated tin compound expressed with at least one sort and R² SnO which are chosen from a group which consists of a compound, an acetylacetone, and ethyl acetate which have a hydroxyl group (however, R² is a univalent hydrocarbon group of the carbon numbers 1-20, and) two or more R² may be the same, or may differ. A reactant produced by making react.

(M-2): (M-1) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.

[Claim 3] The room-temperature-curing nature constituent according to claim 1 whose tin compound (K-1) is a compound expressed with a following formula (4) or a following formula (5).

[Formula 1]



(R³ is a univalent hydrocarbon group of the carbon numbers 1-20 among a formula (4) and (5), and Y is the basis chosen from the group which consists of an amino group, the hydrocarbon group of the carbon numbers 1-8, a halogenated hydrocarbon group, a cyano alkyl group, an alkoxy group, a halogenated alkoxy group, and a cyanoalkoxy group.) two or more R³ may be the same, or may differ, and two or more Y may be the same, or may differ.

[Claim 4] The room-temperature-curing nature constituent according to claim 1 or 2 whose molecular weights of a polymer (A) are 8000-50000.

[Claim 5] The room-temperature-curing nature constituent according to claim 1, 2, or 4 which is a polyoxaalkylene polymer (B) in which a polymer (A) has a hydrolytic silicon group expressed with a formula (1).

[Claim 6] The room-temperature-curing nature constituent according to claim 5 whose molecular-weight-distribution M_w/M_n of a polyoxaalkylene polymer (B) is 1.7 or less.

[Claim 7] A polyoxaalkylene polymer (B) at the end of a polyoxaalkylene polymer produced under existence of an initiator by polymerizing cyclic ether by making a composite metal cyanide complex into a catalyst. The room-temperature-curing nature constituent according to claim 5 or 6 which is a polymer produced by introducing a hydrolytic silicon group expressed with a formula (1).

[Claim 8] A polyoxaalkylene polymer (B) is obtained by polymerizing cyclic ether under existence of an initiator. The room-temperature-curing nature constituent according to claim 5, 6, or 7 which is a polymer produced by molecular-weight-distribution M_w/M_n introducing a hydrolytic silicon group expressed with a formula (1) into an end of a polyoxaalkylene polymer which is 1.7 or less.

[Claim 9] The room-temperature-curing nature constituent according to claim 5, 6, 7, or 8 in which a polyoxaalkylene polymer (B) contains a polymer (D) further produced by polymerizing a polymerization nature unsaturation group content monomer (C) in a polyoxaalkylene polymer (B).

[Claim 10] The room-temperature-curing nature constituent according to claim 9 or 10 which is a monomer which has a hydrolytic silicon group which a part or all of a polymerization nature unsaturation group content monomer (C) has a polymerization nature unsaturation group, and is expressed with a glycidyl group and/or a formula (1).

[Claim 11] The room-temperature-curing nature constituent according to claim 1, 2, 4, 5, 7, 8, 9, 10, or 11 which is a polymer in which some or all of a polymer (A) has simultaneously a hydrolytic silicon group whose a in a hydrolytic silicon group whose a in a formula (1) is 1 or 2, and a formula (1) is 3.

[Claim 12] The room-temperature-curing nature constituent according to claim 1, 2, 4, 5, 7, 8, 9, 10, or 11 which is a polymer in which some or all of a polymer (A) has simultaneously a hydrolytic silicon group whose a in a hydrolytic silicon group whose a in a formula (1) is 1 or 2, and a formula (1) is 3.

[Claim 13] The room-temperature-curing nature constituent according to claim 1, 2, 4, 5, 7, 8, 9, 10, or 11 which is a polymer in which some or all of a polymer (A) has simultaneously a hydrolytic silicon group whose a in a hydrolytic silicon group whose a in a formula (1) is 1 or 2, and a formula (1) is 3.

or 11 in which a polymer (A) contains both polymers which has a hydrolytic silicon group whose a in a polymer in which a in a formula (1) has a hydrolytic silicon group which is 1 or 2, and a formula (1) is 3.

[Claim 14] The room-temperature-curing nature constituent according to claim 1, 2, 4, 5, 7, 8, 9, 10, or 11 whose polymer (A) is a polymer which has only a hydrolytic silicon group whose a in a formula (1) is 3 as a hydrolytic silicon group.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the room-temperature-curing nature constituent which gives the hardened material time to result in an adhesive manifestation excelled [hardened material] in the adhesive property over short and various substrates.

[0002]

[Description of the Prior Art] The method of making an end harden various kinds of polymers which have a hydrolytic silicon group, and using it for sealant, adhesives, etc. is known well, and is a useful method industrially. The polymer especially whose main chain is polyoxyalkylene among such polymers is liquefied at a room temperature, and when a hardened material holds pliability also at low temperature comparatively and uses for sealant, adhesives, etc., it is provided with the desirable characteristic.

[0003] As a constituent which uses such a polymer and it, with a molecular weight of 15000 or less polymer and its constituent are indicated to JP.61-18570.B or JP.61-18582.B with the hydrolytic silicon group content polymer which two hydrolytic bases per silicon atom combine. Such a polymer has insufficient elongation and intensity of a hardened material, and, in the case of the constituent hardened especially with the hygroscopic surface moisture in the air, there was a fault which is remarkably inferior in the hardenability of the depths. Although the polymer in which the amount of polymers and molecular weight distribution are very narrow, and its constituent are known for the compound which has the same end hydrolytic silicon group by JP.3-72527.A, JP.4-283258.A, etc., in this case, although the elongation, intensity, and hardenability of the hardened material have improved, it was not able to be said to make it harden promptly and obtain a hardened material especially, that that hardenability was still enough.

[0004] It is expected that, as for the silicon group which three hydrolytic bases combined with one silicon group, the hydrolysis rate becomes quick as compared with the silicon group which two hydrolytic bases combined, and the cure rate of the polymer which has such an end becomes quick. It is a polymer which has a hydrolytic silicon group which three hydrolytic bases per silicon atom combine with JP.58-10418.B or JP.58-10430.B as such a polymer — a molecular weight \sim 6000 or less — the polymer of low molecular weight is indicated comparatively. Such a polymer was not enough in particular in respect of the inner drying property in low temperature, the elongation of a hardened material, or pliability, although the cure rate was quick to be sure. Especially these polymers had the fault that adhesive strength was low, when it was alone used for the use of adhesives.

[0005] In order to give room-temperature-curing nature to such a polymer, using what is called a curing catalyst is usually performed. As a curing catalyst, organic metallic compounds, such as metal salt of carboxylic acid, acidity, or a basic compound is known, and carboxylate of tin and other organotin compounds are especially common.

[0006]

[Problem(s) to be Solved by the Invention] However, when it is made to harden using as a catalyst tetravalent organotin compounds known as a well-known example, such as dibutyltin dilaurate and dibutyltin diacetate, a cure rate is not what can fully be satisfied, a part for a part far especially from the surface of a cured body — the so-called cure rate of the depths is insufficient, and there was a problem also in an adhesive property with a substrate.

[0007] Although the method of using the reactant of an oxygenated tin compound and an ester

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compound as a curing catalyst was also proposed by JP.1-58219.B as a trial which cancels such a fault, the hardenability in low temperature was not enough.

[0008] In addition, the method of using a dialkyl tin bisacetylacetonate compound as a curing catalyst is also proposed by JP.61-141761.A, and the hardenability in a room temperature and low temperature is also improved. However, combination with the organic polymer known conventionally proposed by the above-mentioned literature etc. was not enough as internal hardenability compared with the speed of hardening of a surface layer.

[0009] A presentation which can improve depths hardenability and an adhesive property with a substrate without worsening the pliability and workability greatly was desired to the polymer which has a hydrolytic silicon group as mentioned above.

[0010]

[Means for Solving the Problem] This invention relates to hardenability, especially a room-temperature-curing nature constituent by which time until it results in an adhesive manifestation was shortened using a specific curing catalyst.

[0011] It is a polymer which has a hydrolytic silicon group expressed with a following formula (1). Some or all of this polymer is a room-temperature-curing nature constituent which uses as an essential ingredient at least one sort of tin compounds (K) chosen from a polymer (A) which is a polymer which has a hydrolytic silicon group whose a of (1) is 3, and a group which consists of following — (K-1) (K-4) as a curing catalyst.

[0012] — SiX_aR¹_{3-a} ... (1)

(R¹ is a univalent organic group substitution of the carbon numbers 1-20, or unsubstituted among a formula (1). X is a hydroxyl group or a hydrolytic basis, and a is 1, 2, or 3.) however, when those R¹ may be the same, or may differ, when two or more R¹ exist, and two or more X exists, those X may be the same or may differ.

[0013] A tin compound (K) : (K-1): A tin compound expressed with a following formula (2).

(K-2): A tin compound expressed with a following formula (3).

(K-3): (K-1) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.

(K-4): (K-2) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.

R²₂Sn (OZ)₂ ... (2)

[R²₂Sn (OZ)]₂ O ... (3)

(R² is a univalent hydrocarbon group of the carbon numbers 1-20 among a formula (2) and (3), and Z is a univalent hydrocarbon group of the carbon numbers 1-20, or an organic group which has a portion which can form a coordinate bond to Sn by intramolecular.) two or more R² may be the same, or may differ, and two or more Z may be the same, or may differ.

[0014] (Polymer (A)) In this invention, although polyoxyalkylene, polyester, polycarbonate, polyolefine, etc. are mentioned as a main chain of a polymer (A), especially a thing that a main chain consists of polyoxyalkylene intrinsically is preferred. Hereafter, a main chain represents and explains a polymer (henceforth a polyoxyalkylene polymer (B)) which is polyoxyalkylene among polymers (A).

[0015] (Polyoxyalkylene polymer (B)) A polyoxyalkylene polymer (B) which has a hydrolytic silicon group expressed with a formula (1) is proposed by JP.3-47825.A, JP.3-72527.A, JP.3-78627.A, etc., for example. As for a polyoxyalkylene polymer (B), it is preferred to use as a raw material a polyoxyalkylene polymer which has a functional group, to make it not pass to the end via an organic group, to introduce a hydrolytic silicon group and to be manufactured so that it may state below.

[0016] As a raw material polyoxyalkylene polymer, a thing of a hydroxyl group end which makes cyclic ether etc. react and is manufactured is preferred under existence of a catalyst and existence of an initiator. As an initiator, a hydroxy compound etc. which have one or more hydroxyl groups can be used. As cyclic ether, ethylene oxide, propylene oxide, butylene oxide, hexyleneoxide, a tetrahydrofuran, etc. are mentioned. As a catalyst, alkaline metal catalysts, such as a potassium system compound and a caesium system compound, a composite metal cyanide complex catalyst, a metalloporphyrin catalyst, etc. are mentioned.

[0017] In this invention, it is preferred to use a polyoxyalkylene polymer of the amount of polymers of

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the molecular weights 8000-50000 as a raw material polyoxyalkylene polymer. Therefore, by [which manufactured using an alkali catalyst etc.] making many halogenated compounds, such as a methylene chloride, react to a polyoxyalkylene polymer of low molecular weight comparatively. It is preferred to use a polyoxyalkylene polymer manufactured using a polyoxyalkylene polymer and a composite metal cyanide catalyst which are acquired by quantifying many.

[0018]Moreover -- especially -- a ratio of weight average molecular weight (M_w) and a number average molecular weight (M_n) -- it being preferred that M_w/M_n uses 1.7 or less polyoxyalkylene polymer, and, as for M_w/M_n , it is still more preferred that it is 1.6 or less, and, as for M_w/M_n , it is preferred that it is especially 1.5 or less.

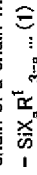
[0019]A polyoxyalkylene polymer (B) which has a hydrolytic silicon group of this invention is obtained by denaturalizing and using an end group as a hydrolytic silicon group further, by using such a polyoxyalkylene polymer as a raw material. When a polyoxyalkylene polymer (B) obtained considering it as a raw material is stiffened so that M_w/M_n of a raw material polyoxyalkylene polymer is small, elongation of a hardened material serves as high intensity greatly, and viscosity of a polymer becomes low, and what has a the same elastic modulus is excellent in workability. Especially in such a polyoxyalkylene polymer, make a composite metal cyanide complex into a catalyst, and Under existence of an initiator, Especially a thing produced by polymerizing alkylene oxide is preferred, and what denaturalized and used an end of such an alkylene oxide polymer as a hydrolytic silicon group is the most preferred.

[0020]A complex which uses zinchexacyano cobaltate as the main ingredients as a composite metal cyanide complex is preferred, and ether and/or an alcoholic complex are especially preferred. The presentation can use what is intrinsically indicated to JP.46-27250.B. In this case, as ether, ethyleneglycol dimethyl ether (glyme), diethylene glycol dimethyl ether (jig lime), etc. are preferred, and especially glyme is preferred from a point of handling at the time of manufacture of a complex. As alcohol, t-butanol is preferred.

[0021]As for a functional group number of a raw material polyoxyalkylene polymer, two or more are preferred. As for especially a functional group number of a raw material polyoxyalkylene polymer, 2 or 3 is preferred to enlarge pliability as the hardened material characteristic. As for especially a functional group number of a raw material polyoxyalkylene polymer, when acquiring a good adhesive property and hardenability, 3-8 are preferred. As a raw material polyoxyalkylene polymer, copolymer of a polyoxyethylene, polyoxypropylene, polyoxy butylene, polyoxy hexylene, polyoxy tetramethylene, and two or more sorts of cyclic ether is specifically mentioned.

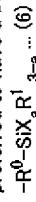
[0022]Especially a desirable raw material polyoxyalkylene polymer is the polyoxypropylene polyol of 2 - 6 value, and are polyoxypropylene diol and polyoxypropylene triol especially. When using for a method of the following (b) or (**), a polyoxyalkylene polymer of olefin ends, such as an allyl end polyoxypropylene monooar, can also be used.

[0023]This polyoxyalkylene polymer (B) has a hydrolytic silicon group expressed with an end or a side chain of a chain with a following formula (1).



(R^1) is a univalent organic group substitution of the carbon numbers 1-20, or unsubstituted among a formula (1), X is a hydroxyl group or a hydrolytic basis, and a is 1, 2, or 3,) however, when two or more R^1 exist, those R^1 may be the same, or may differ, and when two or more X exists, those X may be the same or may differ.

[0024]A hydrolytic silicon group expressed with a formula (1) is usually introduced into a raw material polyoxyalkylene polymer via an organic group. That is, as for a polyoxyalkylene polymer (B), it is preferred to have a basis expressed with a formula (2).



(A divalent organic group, R^1 , X, and a of R^0 are the same as that of the above among a formula (6).)

[0025] R^1 in formula (1) (6) is a univalent organic group substitution of the carbon numbers 1-20, or unsubstituted. It is a with a carbon number of eight or less alkyl group, a phenyl group, or a fluoro alkyl group preferably, and they are a methyl group, an ethyl group, a propyl group, a butyl group, a hexyl group, a cyclohexyl group, a phenyl group, etc. especially preferably, when two or more R^1 exist,

those R^1 may be the same, or may differ.

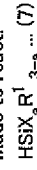
[0026]As a hydrolytic basis in X, a halogen atom, an alkoxy group, an acyloxy group, an alkenyloxy group, a carbamoyl group, an amino group, an aminoxy group, a KETOKISHI mate group, etc. are mentioned, for example.

[0027]As for a carbon number of a hydrolytic basis which has a carbon atom among these, six or less are preferred, and four especially or less are preferred. As desirable X, a with a carbon number of four or less alkoxy group, an alkenyloxy group especially a methoxy group, an ethoxy basis, a propoxy group, or a propenyloxy group can be illustrated, when two or more X exists, those X may be the same or may differ.

[0028]a is 1, 2, or 3. As for the number of hydrolytic silicon groups in one molecule of polymers, 1-8 are preferred, and 2-especially 6 are preferred.

[0029]Although a method in particular of introducing a hydrolytic silicon group to a raw material polyoxyalkylene polymer is not limited, it can be introduced, for example by (b) of the following - a method of (**).

[0030](**) A method to which a hydrosilyl compound expressed with it by a formula (7) after introducing an olefin group into an end of a polyoxyalkylene polymer which has a hydroxyl group is made to react.

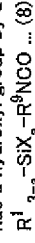


(R^1 , X, and a are the same as the above among a formula (7).)

[0031]A method of making a compound which has an unsaturation group and a functional group react to terminal hydroxyl groups of a polyoxyalkylene polymer which has a hydroxyl group, and combining it by ether bond, ester bond, a urethane bond, or carbonate combination as a method of introducing an olefin group, is mentioned. When polymerizing alkylene oxide, a method of introducing an olefin group into a side chain of a raw material polyoxyalkylene polymer can also be used by adding and carrying out copolymerization of the olefin group content epoxy compounds, such as allyl glycidyl ether.

[0032]When making a hydrosilyl compound react, catalysts, such as a platinum system catalyst, a rhodium system catalyst, a cobalt system catalyst, a palladium system catalyst, and a nickel series catalyst, can be used. Platinum system catalysts, such as chloroplatinic acid, platinum metal, a platinum chloride, and a platinum olefin complex, are preferred. As for a reaction to which a hydrosilyl compound is made to react, it is preferably preferred to carry out at temperature of 60-120 °C for several hours 30-150 °C.

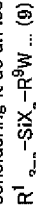
[0033](**) A method to which a compound expressed with an end of a polyoxyalkylene polymer which has a hydroxyl group by a formula (8) is made to react.



(R^1 , X, and a are the same as the above among a formula (8).) R^0 is a divalent hydrocarbon group of the carbon numbers 1-17.

A publicly known urethane-ized catalyst may be used in the case of the above-mentioned reaction. As for the above-mentioned reaction, it is preferably preferred to carry out at temperature of 50-150 °C for several hours 20-200 °C.

[0034](**) A method to which W basis of a silicon compound expressed with a formula (9) to this isocyanate group is made to react after making polyisocyanate compounds, such as tolylene diisocyanate, react to an end of a polyoxyalkylene polymer which has a hydroxyl group and considering it as an isocyanate group end.



(R^1 , R^0 , X, and a are the same as the above among a formula (9).) Active hydrogen containing group as which W is chosen from a hydroxyl group, a carboxyl group, a sulfinyl group, and an amino group (the 1st class or the 2nd class).

[0035](**) A method to which the olefin group and a sulfinyl group of a silicon compound expressed with a formula (9) whose W is a sulfinyl group are made to react after introducing an olefin group into an end of a polyoxyalkylene polymer which has a hydroxyl group.

[0036]W as a silicon compound expressed with a formula (9) which is a sulfinyl group, 3-mercaptopropyltrimethoxysilane, 3-mercaptopropylmethyl dimethoxysilane, 3-mercaptopropyltrimethyl dimethoxysilane, 3-mercaptopropyl triethoxysilane, etc. are mentioned.

[0037] Polymerization initiators, such as a radical generator, may be used, and it may be made to react with radiation or heat in the case of the above-mentioned reaction, without using a polymerization initiator depending on the case. As a polymerization initiator, a polymerization initiator, a metal compound catalyst, etc. of a peroxide system, azo, or a redox system are mentioned, for example. Specifically as a polymerization initiator, 2,2'-azobisisobutyronitrile, 2,2'-azobis 2-methylbutyronitrile, benzoyl peroxide, t-alkyl peroxy ester, acetyl peroxide, diisopropyl peroxy carbonate, etc. are mentioned. As for the above-mentioned reaction, it is preferably preferred to carry out at 50-150 °C for several hours - tens hours 20-200 °C.

[0038] When a main chain is except a polyoxyalkylene polymer) When main chains of a polymer (A) are polyester and polycarbonate, it can manufacture by the same process as a polyoxyalkylene polymer (B) by using polycarbonate of a hydroxyl group end, and polycarbonate of a hydroxyl group end as a raw material, respectively.

[0039] When a main chain is polyolefine, it can manufacture by the same process as a polyoxyalkylene polymer (B) by using polyolefine of hydroxyl group ends, such as polybutadiene polyol and hydrogenation polybutadiene polyol, as a raw material. After having used 1,4-bis(1-chloro-1-methylethyl)benzene as an initiator, making boron trichloride into a catalyst and polymerizing isobutylene, it can manufacture by the same process as a polyoxyalkylene polymer (B) by using a manufactured isobutylene system polymer which carried out the dehydrochlorination reaction and which has an isopropenyl group at the end as a raw material.

[0040] [Hydrolytic silicon group whose a in a formula (1) is 3] It requires that a polymer (A) in this invention is a polymer in which all have "a hydrolytic silicon group whose a in a formula (1) is 3" (henceforth "a hydrolytic silicon group (E)") in part.

[0041] As "a hydrolytic silicon group (E)", especially a basis whose X in a formula (1) is a with a carbon number of four or less alkoxy group, i.e., the Tori alkoxy silyl groups which has a with a carbon number of four or less alkoxy group, is preferred. A polymer which has the Tori alkoxy silyl groups has dramatically high reactivity, and especially its early cure rate is dramatically quick.

[0042] Usually, in a hydrolysis reaction in a hydrolytic silicon group expressed with a formula (1), A silanol group is generated by a reaction with water (silanol group generating reaction expressed with $\text{SiX} + \text{H}_2\text{O} \rightarrow \text{SiOH} + \text{HX}$). It is thought that it progresses by the reaction (condensation reaction) for which the silanol groups furthermore produced condense condensation or a silanol group, and a hydrolytic silicon group, and they produce a siloxane bond. Once a silanol group occurs, it is thought that a condensation reaction goes smoothly. The Tori alkoxy silyl groups has very quick reaction velocity in early stages of a silanol group generating reaction as compared with an alkyl dialkoxy silyl group or dialkyl alkoxy silyl groups. Therefore, it is thought that it has the effect that a hardenability constituent of this invention has short time until it reveals strength property sufficient in a short time and results especially in an adhesive manifestation.

[0043] A direction of the Tori alkoxy silyl groups which has an alkoxy group with a carbon number small among Tori alkoxy silyl groups. Since reaction velocity in early stages of a silanol group generating reaction is quicker than the Tori alkoxy silyl groups which has an alkoxy group with a large carbon number, it is desirable, A trimethoxysilyl group and a triethoxy silyl group are more preferred, and since reaction velocity [in / in a trimethoxysilyl group / early stages of a silanol group generating reaction] is very quick, it is the most desirable. Therefore, it is most preferred that it is a trimethoxysilyl group as "a hydrolytic silicon group (E)". A rate of a hydrolytic silicon group (E) in a hydrolytic silicon group expressed with a formula (1) in a polymer (A) can be changed according to a use, the characteristic to need, etc.

[0044] When a polymer (A) is a polymer which has a hydrolytic silicon group (E) as this hydrolytic silicon group. That is, when about 100% of hydrolytic silicon groups (namely, 80 to 100%) expressed with a formula (1) in a polymer (A) are hydrolytic silicon groups (E), it is effective in a cure rate being large, and a room-temperature-curing nature constituent excellent in especially hardenability that results in an adhesive manifestation is obtained. In this case, it is preferred that 95 to 100% is a hydrolytic silicon group (E) especially 90 to 100% of a hydrolytic silicon group (E) expressed with a formula (1).

[0045] When a hydrolytic silicon group and a hydrolytic silicon group (E) whose a in a formula (1) is 1 or 2 are intermingled, a room-temperature-curing nature constituent which may be compatible in a good extension characteristic and fast curability is obtained.

[0046] In this case, it is preferred that the percentage of a hydrolytic silicon group (E) in all the hydrolytic silicon groups expressed with a formula (1) in a polymer (A) is 5 to 80%. The characteristic according to a demand is freely controllable by changing this rate arbitrarily. That is, when the percentage of a hydrolytic silicon group (E) is 5 to 50%, while raising hardenability, a good extension characteristic and pliability which are needed by sealant etc. can be provided. When the percentage of a hydrolytic silicon group (E) is 50 to 80%, an extension characteristic needed for elastic adhesives etc. can fully be secured, and hardenability can be improved by leaps and bounds.

[0047] As for hydrolytic silicon groups other than a hydrolytic silicon group (E), in a hydrolytic silicon group expressed with a formula (1), it is preferred that a in a formula (1) is especially a hydrolytic silicon group of 2. It is preferred that it is especially a dialkoxy alkyl silyl group which has a with a carbon number of four or less alkoxy group. A dimethoxymethyl silyl group is the most preferred.

[0048] There are following method (**) and (**) in a method of obtaining a polymer (A) that a hydrolytic silicon group which is 1 or 2, and a hydrolytic silicon group (E) were intermingled, for example, and a in a formula (1) may use a method of (**) and (**) together.

(**) As a polymer (A), a in a formula (1) uses a polymer which has simultaneously a hydrolytic silicon group and a hydrolytic silicon group (E) which are 1 or 2.

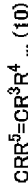
(**) As a polymer (A), a in a formula (1) uses both polymers (A) which has a polyoxyalkylene polymer (B) and a hydrolytic silicon group (E) which have a hydrolytic silicon group which is 1 or 2.

[0049] As for a molecular weight of a polymer (A), although the molecular weight of a polymer (A) in this invention can choose a suitable value according to the use used, it is preferred that it is 8000-50000.

[0050] For uses, such as sealant than to which greater importance is attached to pliability, a polymer of the molecular weights 8000-50000 is preferred. As for a molecular weight, it is preferred that it is especially 8000-25000, and it is most preferred that it is 12000-20000. For a use of adhesives etc. with which intensity is demanded, a polymer of the molecular weights 8000-30000 is preferred. When becoming what has a hardened material weak when lower than 8000 and exceeding 30000, workability gets remarkably bad for hyperviscosity. As for a molecular weight, it is more preferred that it is 8000-20000, and it is preferred that it is especially 12000-20000.

[0051] [Polymer (D) produced by polymerizing a polymerization nature unsaturation group content monomer (C)] A room-temperature-curing nature constituent which uses a polymer (A) as an essential ingredient is excellent in hardenability. In this invention, when using a polyoxyalkylene polymer (B) among polymers (A), it is preferred that a polyoxyalkylene polymer (B) contains a polymer (D) produced by polymerizing a polymerization nature unsaturation group content monomer (C) further. By containing a polymer (D), an effect of adhesive grant in an initial stage of a hardening reaction, i.e., an effect that time until it reveals adhesive strength becomes very short, is acquired.

[0052] Although a compound shown, for example with a following formula (10) is mentioned as a typical thing of a polymerization nature unsaturation group content monomer (C), it is not limited to these.



(R, R³, R⁴, and R⁵ are a hydrogen atom, a halogen atom, or a univalent organic group independently among a formula (10), respectively.)

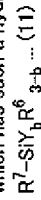
[0053] It is preferred that it is univalent substitution or an unsubstituted hydrocarbon group of the carbon numbers 1-10 as an organic group in R and R⁵. As for R and R⁵, it is more preferred respectively that it is a hydrogen atom. An organic group in R³ and R⁴ Univalent substitution or an unsubstituted hydrocarbon group of the carbon numbers 1-10. It is preferred that they are an alkoxy group, a carboxyl group, an alkoxy carbonyl group, a cyano group, a cyano group content group, an alkenyl group, an acyloxy group, a carbamoyl group, a pyridyl group, a glycidyloxy group, or a glycidyloxy carbonyl group. As for R³, it is preferred that it is especially univalent substitution or an unsubstituted hydrocarbon group of a hydrogen atom, a halogen atom, or the carbon numbers 1-10.

[0054] As an example of a polymerization nature unsaturation group content monomer (C), Styrene monomers, such as styrene, alpha-methylstyrene, and chlorostyrene; Acrylic acid, Methacrylic acid, methyl acrylate, methyl methacrylate, ethyl acrylate, Ethyl methacrylate, butyl acrylate, butyl methacrylate, 2-ethylhexyl acrylate, Acrylic acid, such as 2-ethylhexyl methacrylate, acrylic acid, benzyl, and benzyl methacrylate, Acrylic monomers, such as methacrylic acid or its ester, acrylamide,

and methacrylamide; Acrylonitrile, Cyano group content monomers, such as 2,4-dicyano butene-1; Vinyl acetate, Vinyl ester system monomers, such as vinyl propionate; Butadiene, isoprene, Diene system monomers, such as chloroprene; Vinyl glycidyl ether, allyl glycidyl ether, Glycidyl group content monomer [such as methallyl glycidyl ether, glycidyl acrylate, and glycidyl methacrylate,], and olefins other than these, unsaturation ester species, a halogenation olefin, vinyl ether, etc. are mentioned.

[0055] A polymerization nature unsaturation group content monomer (C) may use only one sort, and may use two or more sorts together. When a cyano group content monomer, a glycidyl group content monomer, or a styrene monomer is used and acrylonitrile, glycidyl acrylate, glycidyl methacrylate, or styrene is used especially, since a further outstanding adhesive property and a mechanical physical property may be revealed, it is desirable. When requiring rubber elasticity especially after hardening, it is preferred to use acrylic ester.

[0056] A polymerization nature monomer which has a hydrolytic silicon group expressed with a formula (1) can be used as a polymerization nature unsaturation group content monomer (C). A compound expressed with a following formula (1) especially as a polymerization nature monomer which has such a hydrolytic silicon group is preferred.



(R^7 is a univalent organic group which has a polymerization nature unsaturation group among a formula (11), R^b is a univalent organic group substitution of the carbon numbers 1-20, or

unsubstituted, Y is a hydroxyl group or a hydrolytic basis, and b is 1, 2, or 3.) however, when two or more R^b exist, those R^b may be the same, or may differ, and when two or more Y exists, those Y may be the same or may differ.

[0057] As a polymerization nature monomer which has a hydrolytic silicon group, a vinyl monomer which has a hydrolytic silicon group, an acrylic monomer which has a hydrolytic silicon group, etc. are mentioned. The following are specifically mentioned and 3-acryloyloxypropyl trimethoxysilane and especially 3-methacryloyl oxypropyl trimethoxysilane are preferred.

[0058] Vinylmethyldimethoxysilane, vinylmethyldiethoxysilane, vinyl methyldi chlorosilane, Vinyltrimethoxysilane, vinyltriethoxysilane, vinyl trichlorosilane, Vinylsilane, such as tris (2-methoxyethoxy) vinylsilane, 3-acryloyloxypropylmethyldimethoxysilane, 3-methacryloyl oxypropyl methyl dimethoxysilane, 3-acryloyloxypropyl trimethoxysilane, 3-acryloyloxypropyl triethoxysilane, Acryloyloxy silanes, such as 3-methacryloyl oxypropyl trimethoxysilane and 3-methacryloyl oxypropyl triethoxysilane, and methacryloyloxy silanes

[0059] A compound which has simultaneously a silicon atom which is a polysiloxane compound which has 2-30 silicon atoms other than these, for example, and was combined with a carbon-carbon double bond and a hydrolytic basis can also be used as a polymerization nature monomer which has a hydrolytic silicon group.

[0060] A polymerization nature monomer which has the above-mentioned hydrolytic silicon group may use only one sort, and may use two or more sorts together. When using a polymerization nature monomer which has a hydrolytic silicon group, this monomer has that preferred of *** for 0.01 ~ 20 weight sections among polymerization nature unsaturation group content monomer (C) 100 weight section.

[0061] As for a part or all of a polymerization nature unsaturation group content monomer (C), it is preferred that it is a monomer which has a hydrolytic silicon group which has a polymerization nature unsaturation group and is expressed with a glycidyl group and/or a formula (1).

[0062] (Polymer composition) When a polyoxyalkylene polymer (B) contains a polymer (D) further, a polymer composition which consists of a polyoxyalkylene polymer (B) and a polymer (D) can be manufactured by a method of (**) shown below - (**).

[0063] (**) How to mix a polymer (D) produced by polymerizing a polymerization nature unsaturation group content monomer (C) beforehand with a polyoxyalkylene polymer (B).

(**) How to polymerize a polymerization nature unsaturation group content monomer (C) in a polyoxyalkylene polymer (B).

(**) How to change an unsaturation group in a polymer group (F) which remains into a hydrolytic silicon group expressed with a formula (1) after polymerizing a polymerization nature unsaturation group content monomer (C) in a polyoxyalkylene polymer group (F) containing an unsaturation group.

A converting method of a method to which a hydrosilyl compound expressed with an unsaturation group by a formula (3) is made to react is preferred.

(**) How to change a precursor into a polyoxyalkylene polymer (B) after polymerizing a polymerization nature unsaturation group content monomer (C) in a precursor of a polyoxyalkylene polymer (B).

(**) How to mix with a polyoxyalkylene polymer (B), rank second if needed, and distill off a solvent or a diluent after polymerizing a polymerization nature unsaturation group content monomer (C) under existence of a solvent or a diluent.

[0064] A solvent can be suitably chosen according to a kind of polymerization nature unsaturation group content monomer (C). As a diluent, an unsaturation group content polyoxyalkylene polymer group (F) is preferred. A polyoxyalkylene polymer group (F) containing an unsaturation group can also be made to exist in a solvent or a diluent in the case of a polymerization.

[0065] Polymerization initiators, such as a radical generator, may be used, and it may be made to polymerize with radiation or heat in the case of a polymerization nature unsaturation group content monomer (C) polymerization, without using a polymerization initiator depending on the case. About a polymerization initiator, polymerization temperature, and polymerization time, it is the same as said (**).

[0066] When using a polymer (D) in this invention, it is preferred that the polyoxyalkylene polymer (B) / polymer (D) is used in the range used as 100 / 1 ~ 1/300 at a weight ratio. It is preferred to be used in 100 / 1 ~ 1/100, further 100 / 1 ~ 1/10 especially in respect of workability etc.

[0067] It may be distributing uniformly in the shape of a particle in a polyoxyalkylene polymer (B), or a polymer (D) may be dissolving uniformly again. When viscosity and workability of a constituent are taken into consideration, it is preferred to distribute uniformly in the shape of a particle.

[0068] In order to reveal an effect of this invention, a compound specific as a curing catalyst is indispensable. When not using such a curing catalyst, reaction velocity of crosslinking reaction of a hydrolytic silicon group will not become sufficient.

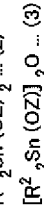
[0069] In this invention, at least one sort of tin compounds (K) chosen from a group which consists of following - (K-1) (K-4) are used as a curing catalyst.

A tin compound (K) : (K-1): A tin compound expressed with a following formula (2).

(K-2): A tin compound expressed with a following formula (3).

(K-3): (K-1) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.

(K-4): (K-2) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.



(R^2 is a univalent hydrocarbon group of the carbon numbers 1-20 among a formula (2) and (3), and Z is a univalent hydrocarbon group of the carbon numbers 1-20, or an organic group which has a portion which can form a coordinate bond to Sn by intramolecular.) two or more R^2 may be the same, or may differ, and two or more Z may be the same, or may differ.

[0070] Or a tin compound (M) which consists of (M-1) and/or (M-2) is used as a curing catalyst. As for a tin compound (K-1) of this invention, it is preferred that it is a tin compound obtained by a method of manufacturing a tin compound (M-1).

A tin compound (M) : (M-1): An oxygenated tin compound expressed with at least one sort and R^2_2 SnO which are chosen from a group which consists of a compound, an acetylacetone, and ethyl acetoacetate which have a hydroxyl group (however, R^2 is a univalent hydrocarbon group of the carbon numbers 1-20, and) two or more R^2 may be the same, or may differ. Reactant produced by making react.

(M-2): (M-1) A mixture or a reactant with a low molecular weight compound (L) which has a hydrolytic silicon group.

[0071] R^2 in an oxygenated tin compound is the same basis as the above-mentioned basis, and specifically A methyl group, An ethyl group, n-propyl group, i-propyl group, n-butyl group, n-amyl

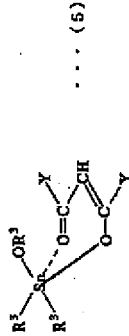
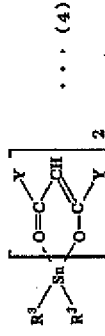
group, i-amyl group, n-hexyl group, a cyclohexyl group, n-octyl group, a 2-ethylhexyl group, a lauryl group, a stearyl group, a phenyl group, etc. are mentioned. As an example of an oxygenated tin compound, although SnO , $(\text{C}_2\text{H}_5)_2\text{SnO}$, $(\text{C}_4\text{H}_9)_2\text{SnO}$, $(\text{C}_6\text{H}_{13})_2\text{SnO}$, $(\text{C}_8\text{H}_{17})_2\text{SnO}$, $(\text{C}_6\text{H}_5)_2\text{SnO}$, etc. are mentioned, for example $(\text{CH}_3)_2$, it is not limited to these.

[0072]As a compound etc. which have a hydroxyl group made to react to an oxygenated tin compound, For example, methanol, ethanol, n-butanol, 2-ethylhexanol,; alcoholic [such as lauryl alcohol,] --- phenols [such as nonyl phenol,]: --- propanolamine. Amino alcohol, such as ethanolamine, dimethyl propanolamine, and diethanolamine; although mercaptoalcohol, such as mercaptoopropanol, etc. are mentioned, it is not limited to these.

[0073]As for a tin compound (K-1), it is preferred that it is especially a compound expressed with a following formula (4) or a following formula (5).

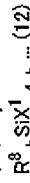
[0074]

[Formula 2]



[0075](Among a formula (4) and (5), R^3 is a univalent hydrocarbon group of the carbon numbers 1-20, and) Y An amino group, a hydrocarbon group (carbon numbers 1-8), a halogenated hydrocarbon group (carbon numbers 1-8). It is the basis chosen from the group which consists of a cyano alkyl group (carbon numbers 1-8 other than a cyano group), an alkoxy group (carbon numbers 1-8), a halogenation alkoxy group (carbon numbers 1-8), and a cyanoalkoxy group (carbon numbers 1-8 other than a cyano group). two or more R^3 may be the same, or may differ, and two or more Y may be the same, or may differ.

[0076]As a low molecular weight compound (L) which has a hydrolytic silicon group used by this invention, a low molecular weight compound which has a hydrolytic silicon group containing a silicon atom which a hydroxyl group and/or a hydrolytic basis combined can be used, and it is preferred that it is 1000 or less molecular weight. A silicon compound especially expressed with a following formula (12) is preferred.



R^8 is a univalent hydrocarbon group substitution of the carbon numbers 1-20, or unsubstituted among a formula, X^1 is a hydroxyl group or a hydrolytic basis, and b is an integer of 0-3. when two or more R^8 exist, it may be the same or may differ, and when two or more X^1 exist, it may be the same or may differ.

[0077] R^8 is a univalent hydrocarbon group substitution of the carbon numbers 1-20, or unsubstituted, and it is with a carbon number of eight or less alkyl group, a phenyl group, or a fluoro alkyl group preferably. They are a methyl group, an ethyl group, a propyl group, a propenyl group, a butyl group, a hexyl group, a cyclohexyl group, a phenyl group, etc. especially preferably.

[0078] X^1 is a hydroxyl group or a hydrolytic basis, and there are a halogen atom, an alkoxy group, an alkoxy group, an amide group, an amino group, an aminoxy group, and a KETOKISHI mate group as a hydrolytic basis, for example. As for especially a carbon number of a hydrolytic basis which has a

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carbon atom among these, four or less are [six or less] preferred. As desirable X, a with a carbon number of four or less lower alkoxy group especially a methoxy group, an ethoxy basis, a propoxy group, etc. can be illustrated.

[0079]Specifically Tetra alkoxy silane; trimethoxy methylsilanes, such as tetraethyl silicate and tetramethyl silicate, Trialkoxy silane, such as TOSI methoxysilane and trimethoxy vinylsilane; Dimethoxy dimethylsilane, Dialkoxy silanes, such as diethoxy dimethylsilane; mono- alkoxy silane, such as methoxy trimethylsilane and ethoxy trimethylsilane, those hydrolyzates, or a partial hydrolyzate is mentioned.

[0080]Those hydrolyzate or partial hydrolyzates, such as acetoxysilane, such as chlorosilanes, such as chloro trimethylsilane and dichlorodimethylsilane, dimethyl diacetoxysilane, and vinyltriacetoxysilane, and N-trimethylsilyl acetamide, are also mentioned.

[0081]Considering influence of an ease of handling, and physical properties on a cured body, alkoxy silane, especially a dialkoxy silane are preferred. Partial condensate of these silicon compounds can also be used.

[0082]a low molecular weight compound (L) and a tin compound (K-1) in which a tin compound (K-3) or (K-4) has a hydrolytic silicon group --- or (K-2) --- from --- it is obtained. It may be a low molecular weight compound (L) which has a hydrolytic silicon group, a tin compound (K-1), or (K-2) a mixture produced by only mixing, and it may react, and may become a reactant, and they may live together.

[0083]As for a tin compound (K-3) or (K-4), it is preferred to obtain by agitating at ordinary temperature -180 ** for 1 to 10 hours in a low molecular weight compound (L) which has a hydrolytic silicon group, a tin compound (K-1), or (K-2) a flask which carried out the nitrogen purge.

[0084]A low molecular weight compound (L) which has a hydrolytic silicon group, a tin compound (K-1), or a using rate of (K-2) can be chosen arbitrarily. In order to improve cold cure nature notably, the range of (L)/(K-1) or (K-2) = 1 / 0.1 - 1/10 is preferred, and especially (L)/(K-1) or (K-2) = 1 / 0.5 - 1/5 are preferred.

[0085]In use of a tin compound (K) as the above-mentioned curing catalyst, it may be used alone, two or more sorts may be used together, and it may use together with other catalysts other than the above. The amount of tin compound (K) used as these curing catalysts is 0.1 - 5 weight section preferably 0.01 to 10 weight section to a total of 100 weight sections of a polymer (A) or a polymer (B). When it becomes less than 0.01 weight sections, pot life is long, but since an adverse effect will appear in physical properties, such as heat resistance, if a cure rate becomes insufficient and ten weight sections are exceeded, it is not desirable.

[0086]Other curing catalysts are used together and a cure rate can be controlled. The following compound is mentioned as such a curing catalyst. A salt, various kinds of acid, and a base substance of carboxylic acid of various metal, such as an alkyl titanate, an organic silicon titanate, and dibutyltin dilaurate, can be used. Specifically, amine salt, such as 2-ethylhexanoic acid tin, 2-ethylhexanoic acid lead and dialkyl tin dicarboxylic acid salt, organic amine, and dibutyl amine 2-ethylhexoate, etc. are mentioned.

[0087](Room-temperature-curing nature constituent) The following additive agent can also be included by room-temperature-curing nature constituent of this invention. Hereafter, an additive agent is explained.

[0088](Bulking agent) A publicly known bulking agent can be used as a bulking agent. Especially the amount of bulking agent used has 50 - 250 preferred weight section 0.001 to 1000 weight section to a total of 100 weight sections of a polymer (A) or a polymer (B), and a polymer (B). The following are mentioned as an example of a bulking agent. These bulking agents may be used independently and may be used together two or more sorts.

[0089]Calcium carbonate which carried out the surface treatment of the surface with fatty acid or a resin acid system organic matter, Colloid calcium carbonate with a mean particle diameter of 1 micrometer or less which impalpable-powderized this calcium carbonate further, Calcium carbonate, such as precipitated calcium carbonate with a mean particle diameter of 1-3 micrometers manufactured with a sedimentation method, and heavy calcium carbonate with a mean particle diameter of 1-20 micrometers. Fumed silica, sedimentation nature silica, a silicic acid anhydride, hydroxyl silicic acids, and carbon black, Magnesium carbonate, diatomite, calcination clay, clay, talc, titanium oxide, bentonite, organic bentonite, ferric oxide, a zinc oxide, an active white, a milt balloon, glass balloons, wood flour, pulp, a cotton chip, mica, and blacking wash farina --- rubbing --- powder

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state bulking agents, such as farina, graphite, aluminum impalpable powder, and the Flint powder. Fibrous fillers, such as asbestos, glass fiber, a glass filament, carbon fiber, the Kevlar textiles, and a polyethylene fiber.

[0090][Plasticizer] A publicly known plasticizer can be used as a plasticizer. The amount of plasticizer used has 0.001 - 1000 preferred weight section to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B). The following are mentioned as an example of a plasticizer.

[0091]Phthalic ester, such as di-(2-ethylhexyl)phthalate (the following, DOP), dibutyl phthalate, and phthalic acid benzyl butyl ester. Aliphatic-carboxylic-acid ester, such as dioctyl adipate, a succinic acid screw (2-methylonyl), dibutyl sebacate, and butyl oleate. Alcohol ester, such as pentaerythritol ester. Phosphoric ester, such as triethyl phosphate and tricresyl phosphate. Epoxy plasticizers, such as chlorinated paraffin. Polyester plasticizers, such as polyester dibasic acid and dihydric alcohol are made to come to react. Polyether, such as polyoxypropylene glycol and its derivative. Polymeric plasticizers, such as oligomer, such as the oligomer of styrene systems, such as Poly alpha-methylstyrene and polystyrene, polybutadiene, Butadiene Acrylonitrile, polychloroprene, polyisoprene, polybutene, hydrogenation polybutene, and epoxidation polybutadiene.

[0092][Adhesive grant agent] An adhesive grant agent is used in order to improve an adhesive property further. As these adhesive grant agents, silane coupling agents, such as (meth)acryloyloxy group content Silang, amino group content Silang, sulfhydryl group content Silang, epoxy group content Silang, and carboxyl group content Silang, are mentioned.

[0093]As (meth)acryloyloxy group content Silang, 3-methacryloyl oxypropyl trimethoxysilane, 3-acryloyloxypropyl trimethoxysilane, 3-methacryloyl oxypropyl methyl dimethoxysilane, etc. are mentioned.

[0094]As amino group content Silang, 3-aminopropyl trimethoxysilane, 3-aminopropyl triethoxysilane, 3-aminopropyl methyl dimethoxysilane, N-(2-aminoethyl)-3-aminopropyl trimethoxysilane, N-(2-aminoethyl)-3-aminopropyl methyl dimethoxysilane, N-(2-aminoethyl)-3-aminopropyl triethoxysilane, 3-ureido propyl triethoxysilane, N-(N-vinylbenzyl 2-aminoethyl)-3-aminopropyl trimethoxysilane, 3-anilino propyl trimethoxysilane, etc. are mentioned.

[0095]As sulfhydryl group content Silang, 3-mercaptopropyl trimethoxysilane, 3-mercaptopropyl triethoxysilane, 3-mercaptopropyl methyl dimethoxysilane, 3-mercaptopropyl methyl diethoxysilane, etc. are mentioned.

[0096]As epoxy group content Silang, 3-glycidyl oxypropyl trimethoxysilane, 3-glycidyl oxypropyl methyl dimethoxysilane, 3-glycidyl oxypropyl triethoxysilane, etc. are mentioned.

[0097]As carboxyl group content Silang, 2-carboxyethyl triethoxysilane, 2-carboxyethyl phenylbis(2-methoxyethoxy)Silang, N-(N-carboxymethyl 2-aminoethyl)-3-aminopropyl trimethoxysilane, etc. are mentioned.

[0098]A reactant produced by making two or more sorts of silane coupling agents react may be used. As an example of a reactant, a reactant of amino group content Silang and epoxy group content Silang, a reactant of amino group content Silang and (meth)acryloyloxy group content Silang, a reactant of epoxy group content Silang and sulfhydryl group content Silang, a reactant of sulfhydryl group content Silang, etc. are mentioned. These reactants are easily obtained by mixing this silane coupling agent and agitating in a room temperature -150 °C temperature requirement for 1 to 8 hours.

[0099]The above-mentioned compound may be used alone and may be used together two or more kinds. The amount of silane coupling agent used has 0 - 30 preferred weight section to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B).

[0100]An epoxy resin may be added as an adhesive grant agent. An epoxy resin hardener may be further used together if needed. A common epoxy resin is mentioned as an epoxy resin which can be added to a constituent of this invention. Specifically, the following can be illustrated. 0 - 100 weight section of the amount used is preferred to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B).

[0101]A bisphenol A-diglycidyl ether type epoxy resin, a bisphenol F-diglycidyl ether type epoxy resin. Fire retardancy type epoxy resins, such as a tetrabromobisphenol A-glycidyl ether type epoxy resin, Novolak type epoxy resin, a hydrogenation bisphenol A type epoxy resin, A glycidyl ether type epoxy resin of a bisphenol A-propylene oxide addition, 4-glycidyl oxy benzoic acid glycidyl, phthalic

acid diglycidyl, Diglycidyl ester system epoxy resins, such as tetrahydrophthalic acid diglycidyl and hexahydrophthalic acid diglycidyl, m-aminophenol series epoxy resin, a diaminodiphenylmethane system epoxy resin, Urethane modified epoxy resin, various cycloaliphatic-epoxy-resin, N, and N-diglycidyl aniline, N,N-diglycidyl o-toluidine, triglycidyl isocyanurate, A vinyl system polymer containing epoxy resins currently generally used and epoxy groups, such as an epoxidation thing of unsaturation polymers, such as glycidyl ether of polyhydric alcohol, such as polyalkylene glycol diglycidyl ether and glycerin, a hydantoin type epoxy resin, and petroleum resin, etc.

[0102]A hardening agent (or curing catalyst) of the above-mentioned epoxy resin may be used together to a constituent of this invention. A hardening agent for epoxy resins generally used as such a hardening agent is mentioned. Specifically, the following can be illustrated. 0.1 to 300 weight section of the amount used is preferred to an epoxy resin.

[0103]Triethylenetetramine, tetraethylenepentamine, diethylamino propylamine, N-aminoethyl piperazine, m-xylene diamine, m-phenylenediamine, Diaminodiphenylmethane, diaminodiphenyl sulfone, isophoronediamine, Amines or those salts, such as 2,4,6-tris(dimethyl aminomethyl) phenol, Or blocked amines, such as a ketimine compound, polyamide resin, Imidazole derivatives, diolamides, and boron trifluoride complex compounds. Phthalic anhydride, a hexahydrophthalic anhydride, a tetrahydrophthalic anhydride, Carboxylic anhydrides, such as a dodecyl succinic acid anhydride and a pyromellitic anhydride, phenoxy resin, carboxylic acid, alcohols, and a polyalkylene oxide system polymer (end amination polyoxypropylene glycol.) that averages a basis which can react to an epoxy group and it has at least one piece in intramolecular Liquefied end functional group content polymers, such as end carboxylation polyoxypropylene glycol, etc. polybutadiene in which an end was embellished with a hydroxyl group, a carboxyl group, an amino group, etc., hydrogenation polybutadiene, an acrylonitrile butadiene copolymer, and an acrylic polymer etc.

[0104][Solvent] When using a constituent of this invention as a hardenability constituent again, a solvent can also be added for the purpose of adjustment of viscosity, and improvement in preservation stability of a constituent. The amount of solvent used has 0.001 - 500 preferred weight section to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B).

[0105]As a solvent, aliphatic hydrocarbon, aromatic hydrocarbon, and halogenated hydrocarbon. Alcohols, ketone, ester species, ether, ester alcohols, ketone alcohol, ether alcohol, ketone ether, ketone ester species, and ester ether can be used. When saving a constituent of this invention at a long period of time, since preservation stability of alcohols improves, they are preferred. As alcohols, alkyl alcohol of the carbon numbers 1-10 is preferred, and methanol, ethanol, isopropanol, isopentyl alcohol, especially hexyl alcohol, etc. are preferred.

[0106][Dehydrator] In order to improve the storage stability of a hardenability constituent of this invention further again, a little dehydrators can be added in the range which has an adverse effect on neither hardenability nor pliability. The amount of dehydrator used has 0.001 - 30 preferred weight section to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B).

[0107]Specifically ORUTOGI acid alkyls, such as methyl orthoformate and ethyl orthoformate, Hydrolytic organic silicon compounds, such as alt-acetic acid alkyls, such as alt-methyl acetate and alt-ethyl acetate, methyl trimethoxysilane, vinyltrimethoxysilane, a tetramethoxy silane, and a tetraethoxysilane, a hydrolytic organic titanium compound, etc. can be used. Vinyltrimethoxysilane and a tetraethoxysilane are preferred especially from a point of cost and an effect.

[0108][Thixotropy grant agent] A thixotropy grant agent may be used again for an improvement of lapet nature. As such a thixotropy grant agent, hydrogenation castor oil, fatty acid amide, etc. are used.

[0109][Antianging agent] As an antiaging agent, an antioxidant generally used, an ultraviolet ray absorbent, and light stabilizer are used suitably again. Each compound of a hindered amine system, a benzotriazol system, a benzophenone series, a benzoate system, a cyanoacrylate system, an acrylate system, a hindered phenol system, the Lynn system, and sulfur systems can be used suitably.

[0110][in addition to this] An air-oxidation hardenability compound and a photoresist compound can be added in order to improve adhesion and surface tackiness of a paint over a long period of time again. The amount of air-oxidation hardenability compound used to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B) 0.001 - 50 weight section. The amount of photoresist compound used has 0.001 - 50 preferred weight section to a total of 100 weight sections of a polymer (A) or a polymer (A), and a polymer (B).

[0111] Drying oil represented by tung oil, linseed oil, etc. as such an air-oxidation hardenability compound. An acrylic polymer which denaturalized with various alkyl resins produced by denaturalizing this compound, and drying oil. Various denaturation things (maleinized denaturation, boiled oil denaturation, etc.) of diene system polymers, such as a polymer of silicone resin, polybutadiene, and diene of the carbon numbers 5-8 and a copolymer, and also this polymer, or a copolymer, etc. are mentioned. As a photorealist compound, polyfunctional acrylate is usually used. In addition, organic colors, such as inorganic pigments, such as iron oxide, chrome oxide, and titanium oxide, and copper phthalocyanine blue, and Phthalocyanine Green, are mentioned to paints.

[0112] A room-temperature-curing nature constituent of this invention is suitable for a use as which can be used for sealant, a water blocking material, adhesives, a coating agent, etc., and dynamic flattery nature to sufficient cohesive force and adherend of especially the hardened material itself is required.

[0113]

[Example 1] The example and comparative example which produced the hardened material are shown below using the polymer (P1-P11, P13-P23) manufactured in the examples 1-11, 13-23 of manufacture. A part shows a weight section. In the examples 1-12 of manufacture, a hydroxyl value conversion molecular weight shows the molecular weight converted from the hydroxyl value of the polyoxyalkylene polymer which has a hydroxyl group which is a raw material. M_w/M_n is the value measured by the gel permeation chromatograph using a tetrahydrofuran as a solvent. The analytical curve was prepared using polystyrene.

[0114] (Example 1 of manufacture) Glycerin was used as the initiator and the bottom propylene oxide of existence of a zinchexacyano cobaltate glyme complex compound catalyst was made to react. The methanol solution of sodium methoxide was added to polyoxypropylene triol of hydroxyl value conversion molecular weight 17000 and $M_w/M_n=1.3$, scale loss pressing-down methanol was distilled off, and the terminal hydroxyl groups of polyoxypropylene triol was changed into sodium alcoholate. Next, the allyl chloride was made to react. The unreacted allyl chloride was removed and refined and allyl group end polypropylene oxide was obtained (let this be the polymer U1.). When the hydroxyl group which remains was analyzed by the measuring method of the hydroxyl value, they were 0.01 millimol/g. The trimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst to the polymer U1., and the polymer P1 which has an average of 2.0 trimethoxysilyl groups at the end was obtained.

[0115] (Example 2 of manufacture) The polyoxypropylene glycol of hydroxyl value conversion molecular weight [which used propylene glycol as the initiator, made the bottom propylene oxide of existence of a zinchexacyano cobaltate glyme complex compound catalyst react, and was obtained] 17000, and $M_w/M_n=1.3$ is used. The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained (the hydroxyl groups which remain are 0.01 millimols/g). The trimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst to this reactant, and the polymer P2 which has an average of 1.3 trimethoxysilyl groups at the end was obtained.

[0116] (Example 3 of manufacture) Polyoxypropylene hexaol of hydroxyl value conversion molecular weight [which used sorbitol as the initiator, made the bottom propylene oxide of existence of a zinchexacyano cobaltate glyme complex compound catalyst react, and was obtained] 15000, and $M_w/M_n=1.3$ is used. The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained (the hydroxyl groups which remain are 0.01 millimols/g). The trimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst to this reactant, and the polymer P3 which has an average of 3.9 trimethoxysilyl groups at the end was obtained.

[0117] (Example 4 of manufacture) The mixture which made the mole ratio methyl dimethoxysilane and trimethoxysilane which are hydrosilyl compounds, and was mixed in proportion of 30 to 70 is made to react under existence of a platinum catalyst to the polymer U1 manufactured in the example 1 of manufacture. The polymer P4 which has simultaneously an average of 0.6 methyl dimethoxy silyl group and an average of 1.4 trimethoxysilyl groups at the end was obtained.

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[0118] (Example 5 of manufacture) 3-mercaptopropyltrimethoxysilane which is a silyl compound to the polymer U1 manufactured in the example 1 of manufacture, it was made to react using the 2,2'-azobis 2-methylbutyronitrile which is a polymerization initiator, and the polymer P5 which has an average of two trimethoxysilyl groups at the end was obtained.

[0119] (Example 6 of manufacture) It refined, after polymerizing propylene oxide using the zinchexacyano cobaltate catalyst by having used glycerin as the initiator and obtaining polyoxypropylene triol of hydroxyl value conversion molecular weight 17000 and $M_w/M_n=1.3$.

Isoocyanate propyltrimethoxysilane was added to this, the urethane-ized reaction was performed, the end was changed into the trimethoxysilyl group, and the polymer P6 of the molecular weight 18000 which has an average of two trimethoxysilyl groups at the end was obtained.

[0120] (Example 7 of manufacture) To the polymer U1 manufactured in the example 1 of manufacture, methyl dimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst, and the polymer P7 which has an average of two methyl dimethoxy silyl groups at the end was obtained.

[0121] (Example 8 of manufacture) The polyoxypropylene glycol of hydroxyl value conversion molecular weight [which used propylene glycol as the initiator, made the bottom propylene oxide of existence of a zinchexacyano cobaltate glyme complex compound catalyst react, and was obtained] 7000, and $M_w/M_n=1.2$ is used. The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained (the hydroxyl groups which remain are 0.01 millimols/g). Methyl dimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst to this reactant, and the polymer P8 which has an average of 1.3 methyl dimethoxy silyl groups at the end was obtained.

[0122] (Example 9 of manufacture) The polyoxypropylene glycol of hydroxyl value conversion molecular weight [which used propylene glycol as the initiator, made the bottom propylene oxide of existence of a zinchexacyano cobaltate glyme complex compound catalyst react, and was obtained] 7000, and $M_w/M_n=1.2$ is used. The polypropylene oxide which has an allyl group at the end by the same method as the example 1 of manufacture was obtained (the hydroxyl groups which remain are 0.01 millimols/g). The trimethoxysilane which is a hydrosilyl compound was made to react under existence of a platinum catalyst to this reactant, and the polymer P9 which has an average of 1.3 trimethoxysilyl groups at the end was obtained.

[0123] (Example 10 of manufacture) The methanol solution of sodium methoxide was added to polyoxypropylene diol of the hydroxyl value conversion molecular weight 3000 obtained using the potassium hydroxide catalyst, scale loss pressing-down methanol was distilled off, and terminal hydroxyl groups was changed into sodium alcoholate. Next, the allyl chloride was made to react continuously, after making it react to chlorobromomethane and performing polymers quantification. The unreacted allyl chloride was removed and refined and the polypropylene oxide ($M_w/M_n=1.9$) which has an allyloxy group at the end was obtained (the hydroxyl groups which remain are 0.01 millimols/g). The polymer P10 of the molecular weight 7000 which makes the trimethoxysilane which is a hydrosilyl compound react to this under existence of a platinum catalyst, and has an average of 1.3 trimethoxysilyl groups at the end was obtained.

[0124] (Example 11 of manufacture) Polyoxypropylene diol of hydroxyl value conversion molecular weight [which was obtained using the potassium hydroxide catalyst] 6000 and $M_w/M_n=1.9$ is used. The polypropylene oxide which has an allyloxy group at the end by the same method as the example 1 of manufacture was obtained (the hydroxyl groups which remain are 0.01 millimols/g). The trimethoxysilane which is a hydrosilyl compound was made to react to this under existence of a platinum catalyst, and the polymer P11 which has an average of 1.3 trimethoxysilyl groups at the end was obtained.

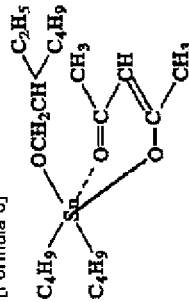
[0125] (Example 12 of manufacture) Add 49.8 g (0.2 mol) of dibutyltin oxide to toluene 150cm³ in glass reactors, and 13.0 g (0.1 mol) of 2-ethylhexanol is added, it was made to react removing the water which carries out azeotropy to bottom toluene of heating churning until the water of the amount of theories finishes distilling off. 10.0 g (0.1 mol) of acetylacetones are added after that, and it was made to react removing the water which carries out azeotropy to toluene further until the water of the amount of theories finishes distilling off. It filtered in order to remove a little sediments, and it distilled off under decompression of toluene further, and the light yellow fluid [compound (Q)] was

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obtained. It checked that the compound expressed with ** 3 was generating.

[0126]

[Formula 3]



[0127](Examples 1-10 and comparative examples 1-4) The inside of the polymers P1-P11, Calcium carbonate to 100 copies of polymers shown in Tables 1 and 2 150 copies. After adding 50 copies and a thixotropy grant agent for di-2-ethylhexyl phthalate, adding one copy for three copies and a phenolic antioxidant and kneading under a nitrogen atmosphere. Two copies or dibutyltin bisacetylacetonate (AC) 2 copy was added, the compound (Q) obtained in the example 12 of manufacture as a curing catalyst was kneaded further, and the hardenability constituent was obtained. However, the comparative example 2 and the comparative example 4 used two copies of dibutyltin dilaurate (DBTDL) instead of a tin compound (Q) or dibutyltin bisacetylacetonate (AC) using 100 copies of things mixed by five P1/of examples P7=7 / 3 (weight ratio). The result of having done the following examinations about the obtained hardenability constituent is shown in Table 1 and Table 2.

[0128]H type tensile test sample was produced based on <50% tensile stress and adhesive failure state> JIS A5758, using an aluminum plate as adherend. After recuperating oneself for 14 days at 30 more ** for 14 days according to a normal condition, the tensile test was done and the fracture state with 50% tensile stress and the substrate at the time of destruction was measured. As a fracture state, subsequently, thin layer cohesive failure (TCF) is preferred, and since the adhesive property is insufficient, interfacial failure (AF) is not preferred [cohesive failure (GF) is the most preferred, and / cohesive failure].

[0129]The hardenability constituent was slushed so that it might become a thickness of 4 cm into the cup with a <penetration> diameter of 4 cm of a cylindrical shape, and it was neglected under the atmosphere of 65% humidity at 20 ** for 6 hours. The situation of hardening to a depth direction from the surface was seen using the needle for [1.25g.] asphalt using the penetrator based on JISK2530 after that. That is, the degree of penetration of the needle for 5 seconds from the perpendicular direction upper part to a lower part (penetration, unit:cm) was measured. The one where penetration is larger means that hardening from the surface is not progressing.

[0130](Example 13 of manufacture) Based on the method indicated to JP,1-170681,A, 1,4-bis(1-chloro-1-methylethyl)benzene is used as an initiator. The molecular weight which has an isopropenyl group at a rate of about 92% in the both ends which polymerized isobutylene by making boron trichloride into a catalyst, and which carried out back dehydrochlorination and were manufactured makes chloroplatinic acid with a catalyst the isobutylene system polymer of about 5000, and trichlorosilane is made to react in 90 ** 12 hours. The polyisobutylene system polymer P13 which has a trimethoxysilyl group at the end by making methanol react to methyl orthoformate furthermore was obtained.

[0131](Example 14 of manufacture) Based on the method indicated to JP,1-170681,A, 1,4-bis(1-chloro-1-methylethyl)benzene is used as an initiator. The molecular weight which has an isopropenyl group at a rate of about 92% in the both ends which polymerized isobutylene by making boron trichloride into a catalyst, and which carried out back dehydrochlorination and were manufactured makes chloroplatinic acid with a catalyst the isobutylene system polymer of about 5000, and methylchlorosilane is made to react in 90 ** 12 hours. The polyisobutylene system polymer P14 which has a methyl dimethoxy silyl group at the end by making methanol react to methyl orthoformate furthermore was obtained.

[0132](Example 15 of manufacture) 90-mol% of 3-isocyanate propyltrimethoxysilane was made to react to an end to the terminal hydroxyl groups of the hydrogenation polybutadiene (polytail HA, Mitsubishi Chemical make) which has a hydroxyl group, and the hydrogenation polybutadiene

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polymerization object P15 which has a trimethoxysilyl group at the end was acquired. [0133](Example 16 of manufacture) the hydrogenation polybutadiene (polytail HA) which has a hydroxyl group at the end 90-mol% of 3-isocyanate propylmethyl dimethoxysilane was made to react to the Mitsubishi Chemical terminal hydroxyl groups, and the hydrogenation polybutadiene polymerization object P16 which has a methyl dimethoxy silyl group at the end was acquired. [0134](Examples 11-14 and comparative examples 5-7) As opposed to 100 copies of polymers shown in Table 3 among the polymers P13-P16, 100 copies and heavy calcium carbonate for fatty acid surface treatment calcium carbonate 50 copies, As a plasticizer a high boiling point hydrocarbon system plasticizer (high ZORU of Nippon Oil Chemicals) 75 copies, After having added three copies and five copies of sodium sulfate hydrates for the thixotropy grant agent, having added one copy for the phenolic antioxidant, adding one copy and one copy of ultraviolet ray absorbent for the 3rd class hindered amine light stabiliser and kneading under a nitrogen atmosphere, two copies of curing catalysts shown in a table were added, it kneaded further, and the hardenability constituent was obtained. However, 14 used 100 copies of things mixed by P13/of examples P14=1 / 1 (weight ratio). [0135]The obtained hardenability constituent was used and the same examination as Examples 1-10 and the comparative examples 1-4 was done. A result is shown in Table 3.

[0136](Example 17 of manufacture) 100 g of the polymer P1 was put into the reactor with a churning group, and it heated at 100 **, and it added over 3 hours from the dropping funnel there, agitating the solution of 15 g of acrylonitrile, 15g of styrene, and 0.3 g of 2,2'-azobisisobutyronitrile. After continuing heating churning at 100 ** for further 2 hours, devolatilization operation was performed under decompression and the polymer mixture P17 of the letter of nebula was obtained.

[0137](Example 18 of manufacture) The polymer mixture P18 was manufactured like the example 17 of manufacture except using the polymer P7 (methyl dimethoxy silyl group end) for a raw material instead of the polymer P1 (trimethoxysilyl group end).

[0138](Example 19 of manufacture) 100 g of the polymer P1 was put into the reactor with an agitator, and 50 g of toluene was added and diluted. Heat this mixture at 100 ** and 20 g of acrylonitrile, 20 g of styrene. The solution which dissolved 0.3 g of 2,2'-azobisisobutyronitrile in 5g of glycidyl methacrylate and 2 g of 3-methacryloyl oxypyl trimethoxysilane was dropped under churning over 3 hours. The toluene solution of 0.2 g of 2,2'-azobisisobutyronitrile was dropped at the end of dropping back pan over 30 minutes, and the back carried out heating churning at 100 ** for 3 hours. Toluene was distilled out of the obtained mixture under 100 ** decompression, and the polymer mixture P19 of the letter of nebula was obtained.

[0139](Example 20 of manufacture) The polymer mixture P20 of the letter of nebula was manufactured like the example 19 of manufacture except replacing a raw material with the polymer P1 (trimethoxysilyl group end), and using the polymer P7 (methyl dimethoxy silyl group end).

[0140](Example 21 of manufacture) 100 g of the polymer P1 was put into the reactor with an agitator. Heat this at 100 ** and 5 g of styrene, 10 g of methyl methacrylate. The solution which dissolved 7 g of butyl methacrylate, 2 g of methacrylic acid octadecyl, 18 g of 3-methacryloyl oxypyl trimethoxysilane, and 0.5 g of 2,2'-azobisisobutyronitrile was dropped under churning over 3 hours. The toluene solution of 0.2 g of 2,2'-azobisisobutyronitrile was dropped at the end of dropping back pan over 30 minutes, and the back carried out heating churning at 100 ** for 3 hours. Toluene was distilled out of the obtained mixture under 100 ** decompression, and the polymer mixture P21 of the letter of nebula was obtained.

[0141](Example 22 of manufacture) The polymer P22 was manufactured like the example 21 of manufacture except replacing a raw material with the polymer P1 (trimethoxysilyl group end), and using the polymer P7 (methyl dimethoxy silyl group end).

[0142](Examples 15-19 and comparative examples 8-11) The inside of the polymers P17-P22, Fatty acid surface treatment calcium carbonate to 100 copies of polymers shown in Table 4 50 copies. After having added one copy for the phenolic antioxidant, adding one copy and one copy of ultraviolet ray absorbent for the 3rd class hindered amine light stabiliser and kneading under a nitrogen atmosphere, two copies of curing catalysts shown in a table were added, it kneaded further, and the hardenability constituent was obtained. However, 19 used 100 copies of things mixed by P17/of examples P18=1 / 1 (weight ratio).

[0143]The same examination as Examples 1-10 and the comparative examples 1-4 was done. A result is shown in Table 4.

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[0144]

[Table 1]

	実施 例1	実施 例2	実施 例3	実施 例4	実施 例5	実施 例6	実施 例7	実施 例8	実施 例9	実施 例10
重合体	PI	P2	P3	P4	P5	P6	P7	P8	P9	P10
硬化性 樹脂	AC	AC	AC	Q	Q	Q	Q	Q	AC	Q
50%引張 応力 (kg/cm ²)	2.8	3.3	3.3	2.4	2.5	2.4	3.0	4.5	3.2	3.5
機械的 強度 (CF /TCF/AF)	100/0 /0	100/0 /0	100/0 /0	56/5/0	95/5/0	100/0 /0	100/0 /0	90/10/0	70/20 /20	30/ 20/0
針入度 (cm)	0.3	0.2	0.2	0.5	0.5	0.5	0.4	0.4	0.7	0.9

[0145]

[Table 2]

	比較 例1	比較 例2	比較 例3	比較 例4
重合体	P7	P1	P8	P1
硬化性樹脂	AC	DETDL	AC	DETDL
50%引張 応力 (kg/cm ²)	2.3	2.1	4.0	2.5
機械的 強度 (CF /TCF/AF)	90/0 /10	20/10 /70	30/0 /20	50/20 /30
針入度 (cm)	2.2	3.8	2.7	2.8

[0146]

[Table 3]

	実施 例11	実施 例12	実施 例13	実施 例14	比較 例5	比較 例6	比較 例7
重合体	P13	P15	P15	P13/P14	P13	P14	P16
硬化性樹脂	AC	AC	Q	AC	DETDL	AC	AC
50%引張 応力 (kg/cm ²)	3.1	2.5	2.4	2.8	2.6	2.5	2.2
機械的 強度 (CF /TCF/AF)	100/0/ 0	100/0/ 0	100/0/ 0	100/0/ 0	40/20/ 40	20/10/ 70	15/10/ 75
針入度 (cm)	0.6	0.7	0.7	1.5	3.8	2.5	2.5

[0147]

[Table 4]

	実施 例15	実施 例16	実施 例17	実施 例18	実施 例19	比較 例8	比較 例9	比較 例10	比較 例11
重合体	P17	P19	P21	P21 18	P17/P18	P17	P18	P20	P22
硬化性樹脂	AC	AC	AC	Q	DETDL	DETDL	AC	AC	Q
50%引張 応力 (kg/cm ²)	5.5	4.5	5.0	5.0	4.6	5.2	3.5	3.2	3.4
機械的 強度 (CF /TCF/AF)	100/0 /0	100/0 /0	100/0 /0	95/5/ 0	40/20 /40	20/10 /70	15/10 /70	15/10 /70	15/10 /70
針入度 (cm)	0.5	0.8	0.8	0.9	3.0	18.5	7.5	6.9	6.5

[0148]

[Effect of the Invention]The hardenability of the depths is improved and the hardenability constituent in this invention has the effect that an adhesive property with a substrate is good.

[Translation done.]